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Division of Multidisciplinary Chemistry - Molecular Rheology -

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University of Minnesota, USA, 16 January 2009
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Suranaree University of Technology, Thailand, 13–14 March 2009
Illinois Institute of Technology, USA, 16–20 May 2009
Shanghai Jiao Tong University, China, 5–6 November 2009
Suranaree University of Technology, Thailand, 1 November 2008–31 October 2009

Scope of Research

The molecular origin of various rheological properties of material is studied. Depending on time and temperature, homogeneous polymeric materials exhibit typical features of glass, rubber, and viscous fluid while heterogeneous polymeric systems exhibit plasticity in addition to these features. For a basic understanding of the features, the molecular motion and structures of various scales are studied for polymeric systems in deformed state. Measurements are performed of rheological properties with various rheometers, of isochronal molecular orientation with flow birefringence, and of auto-correlation of the orientation with dynamic dielectric spectroscopy. Direct observation of molecular motion is also carried out with fluorescent microscopy and molecular simulations.

Research Activities (Year 2009)

Publications

Watanabe H: Slow Dynamics in Homopolymer Liquids, *Polym. J.*, **41**(11), 929–950 (2009).

Masubuchi Y, Furuichi K, Horio K, Uneyama T, Watanabe H, Ianniruberto G, Greco F, Marrucci G: Primitive Chain Network Simulations for Entangled DNA Solutions, *J. Chem. Phys.*, **131**, [114906-1]-[114906-8] (2009).

Kawakita H, Uneyama T, Kojima M, Morishima K, Masubuchi Y, Watanabe H: Formation of Globules and Aggregates of DNA Chains in DNA/Polyethylene Glycol/monovalent Salt Aqueous Solutions, *J. Chem. Phys.*, **131**, [094901-1]-[094901-9] (2009).

Matsumiya Y, Inoue T, Iwashige T, Watanabe H: Dielectric Relaxation of Polymer/Carbon Dioxide Systems,

Macromolecules, **42**, 4712–4718 (2009).

Qiao X, Li W, Watanabe H, Sun K, Chen X: Rheological Behavior of Biocomposites of Silk Fibroin Fiber and Poly(ϵ -caprolactone): Effect of Fiber Network, *J. Polym. Sci. B: Polym. Phys.*, **47**, 1957–1970 (2009).

Okuda S, Inoue Y, Masubuchi Y, Uneyama T, Hojo M: Wall Boundary Model for Primitive Chain Network Simulations, *J. Chem. Phys.*, **130**, [214907-1]-[214907-7] (2009).

Sato H, Masubuchi Y, Watanabe H: DNA Diffusion in Aqueous Solution in Presence of Suspended Particles, *J. Polym. Sci. B Polym. Phys.*, **47**, 1103–1101 (2009).

Uneyama T, Masubuchi Y, Horio K, Matsumiya Y, Watanabe H, Pathak JA, Roland CM: A Theoretical Analysis of Rheodielectric Response of Type-A Polymer

DNA Diffusion in Aqueous Solution in Presence of Suspended Particles

Although nano-particles, which are comparable in size to polymer chains, are widely used as fillers to polymer matrixes for developing functional and high performance materials, the dynamics of polymers constrained between solid particles has not been well elucidated. In this study, dynamics of individual polymer under such condition was investigated with fluorescent microscopy using DNA solutions as model systems as shown in Figure 1.

For individual T4 and λ DNA molecules in aqueous suspensions of spherical polystyrene particles with diameter of 1 μm , it was found that i) the radius of gyration of DNA is independent of the particle volume fraction, ϕ_p ,

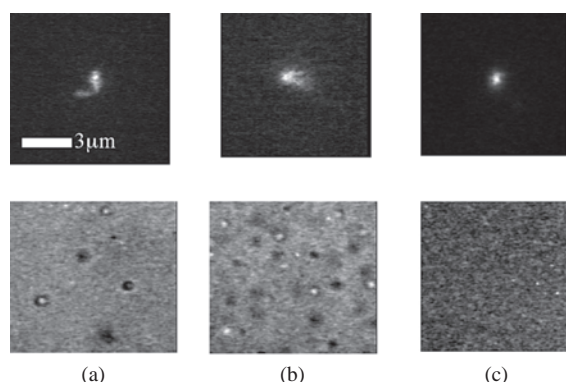


Figure 1. Typical snapshots of the probe T4-DNA (upper photos) and the particles (lower photos) at various particle volume fractions of (a) 4.9×10^{-4} , (b) 4.9×10^{-3} and (c) 9.8×10^{-2} .

Chains, *J. Polym. Sci. B Polym. Phys.*, **47**, 1039-1057 (2009).

Moriya M, Roschztardt F, Nakahara Y, Saito H, Masubuchi Y, Asakura T: Rheological Properties of Native Silk Fibroins from Domestic and Wild Silkworms, and Flow Analysis in Each Spinneret by a Finite Element Method, *Biomacromolecules*, **10**, 929-935 (2009).

Uneyama T: Coarse-Grained Brownian Dynamics Simulations for Symmetric Diblock Copolymer Melts Based on the Soft Dumbbell Model, *Nihon Reorogi Gakkaishi (J. Soc. Rheol. Japan)*, **37**, 81-90 (2009).

Grants

Watanabe H, Creation of Non-equilibrium Soft Matter Physics: Structure and Dynamics of Mesoscopic Systems, Grant-in-Aid for Scientific Research on Priority Areas, October 2006–March 2011.

Masubuchi Y, Multi-scale Simulations for Soft Matters, Core Research for Evolutional Science and Technology, Japan Science and Technology Agency, October 2006–

March 2012.

ii) DNA diffusion is not sensitive to ϕ_p up to a certain critical ϕ_p where the average distance between particle surfaces is close to DNA size, and iii) the DNA diffusion becomes slower at higher ϕ_p . The diffusion coefficient of DNA was larger, by a factor of 2, in the suspensions at intermediate ϕ_p than in the corresponding confined geometry (channel/slit between fixed walls), while this difference asymptotically vanished with increasing ϕ_p (see Figure 2). This result suggested that the DNA diffusion in the suspensions with intermediate ϕ_p is accelerated by the particle motion. In fact, the diffusion coefficient measured for DNA in the suspensions was semi-quantitatively described by the Rouse constraint-release model considering the matrix effect on the probe chain diffusion.

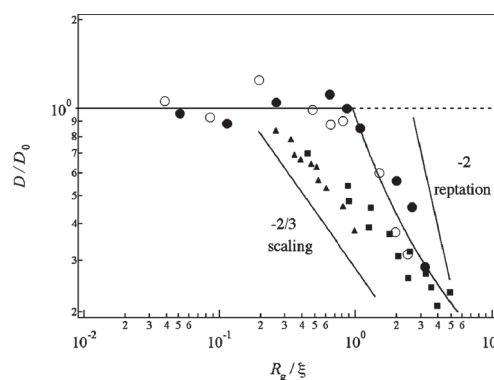


Figure 2. Normalized diffusion constant plotted against the normalized characteristic length of the constraint. Filled and unfilled circles are for T4 and λ DNA, respectively. Square and triangle are data for DNA in confined geometries. Solid lines are for the blob scaling theory and the reptation theory. Solid curves are prediction of the Rouse constraint-release model for T4 DNA. Horizontal broken line shows $D/D_0=1$.

March 2012.

Matsumiya Y, Dynamics of Ionic Liquids in Polymer Networks, Grant-in-Aid for Young Scientists (B), April 2007–March 2009.

Masubuchi Y, A Novel Molecular Model for Branched Polymer Dynamics, Grant-in-Aid for Scientific Research (B), April 2008–March 2011.

Watanabe H, Effect of Thermodynamical and Geometrical Constraints on the Dynamics of Block-copolymers, Grant-in-Aid for Scientific Research (B), April 2009–March 2012.

Awards

Masubuchi Y, 2009 Award of Molecular Simulation Society of Japan.

Uno A, The Best Presentation Award in the 2009 Annual Meeting of the Society of Rheology, Japan.

Hiramoto K, The Best Presentation Award in the 2009 Autumn Meeting of the Society of Rheology, Japan.